Extended Abstracts of the 2020 International Conference on Solid State Devices and Materials, VIRTUAL conference, 2020, pp589-590

Distributed Feedback Laser with Methyl Ammonium Lead Bromide Embedded in Channel-Type Waveguides

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Abstract

$CH_3NH_3PbBr_3$ distributed feedback (DFB) lasing in channel-type waveguides fabricated by a focused ion beam lithography on SiO_2/Si substrate is demonstrated. The highly oriented $CH_3NH_3PbBr_3$ crystals grown inside the waveguide with the DFB gratings show splitting narrow peaks with ~0.8 nm linewidth above a threshold excitation density of 47 μJ/cm^2 which is one order of magnitude lower than that for amplified spontaneous emission in the non-DFB laser. The finite-differential time-domain calculations well reproduce the two emission peaks at λ = 545.5 and 548 nm (two standing waves) corresponding to the DFB modes dephased by π/2 for the second-order diffraction.

1. Introduction

Organometal halide perovskite materials ($CH_3NH_3PbX_3$) have been intensively investigated in the field of optoelectronic devices owing to their exceptional electronic and optical properties. The features of the perovskites having high absorption coefficients, excellent charge mobilities [1] and long diffusion lengths [2] as well as low cost solution processability are important for photonic devices, like light-emitting diodes (LEDs) [3], light-emitting transistors (LETs) [4], light-emitting electrochemical cells (LECs) [5], and lasers [6-8]. To realize lasing at a low excitation threshold, various types of cavity structures have been designed such as vertical-cavity surface-emitting lasers (VCSELs) with distributed Bragg reflectors (DBRs) [9], whispering-gallery-mode (WGM) [10] and axial Fabry-Pérot resonators [11]. Distributed feedback (DFB) perovskite lasers which allow emission wavelength control, beam direction, and mode selection have also been recently reported [7, 12].

In our previous work, perovskite VCSELs and LEDs fabricated by using a simple solution process called 'cast-capping' method [13] have been demonstrated. We have also reported WGM lasing from size-controlled $CH_3NH_3PbBr_3$ crystals grown in a quartz microcapillary [10]. Although single mode lasing has been obtained in the microcapillary with a diameter of 2 μm [10], the lasing wavelength is not arbitrarily controllable. Moreover, there is a challenge to introduce electrodes inside the microcapillary to fabricate LEDs. In the present study, we have modified the microcapillary method by replacing it with channel-type waveguides having DFB gratings. In this report, we fabricated the DFB gratings using a focused ion beam (FIB).

2. Fabrication of perovskite crystals and DFB gratings

30wt.% solutions of methylammonium bromide (CH_3NH_3Br, Wako, 98 %) and lead bromide ($PbBr_2$, Wako, 99.999 %) in dimethyl sulfoxide (DMSO, Wako) were blended at a molar ratio of 1:1 to prepare the perovskite precursor solution. To form $CH_3NH_3PbBr_3$ crystals inside the channel-type waveguides, the perovskite precursor solution was spin-coated on substrates at a speed of 5000 rpm for 30 s in an N_2-filled glove box, followed by thermal annealing at 40 °C for 24 h. The formation of perovskite films on the SiO_2/Si substrate without DFB structure was carried out under similar conditions as a reference.

A schematic representation of a channel-type waveguide (10 μm width) with DFB structure is shown in Fig. 1(a). For the fabrication of DFB resonators, FIB etching was performed to engrave 2nd-order gratings in the channel-type waveguides at an acceleration voltage of 40 kV and an aperture with a diameter of 80 μm. Taking into account the lasing wavelength in the perovskite crystals, the effective refractive index of the perovskite [14], and the technical limitations (grating periods and depths) of FIB milling process, the DFB gratings with a period of 252 nm were fabricated.

An atomic force microscope (AFM, Hitachi E-sweep) image of the fabricated DFB gratings inside the channel-type waveguides is shown in Fig. 1(b). It was found from the cross-sectional profile of the gratings that the DFB resonator was fabricated by FIB processing as designed (periods: 253 nm, depths: 60 nm). Moreover, the strong (100) and (200) reflections appearing in its X-ray diffraction (XRD) pattern showed that highly oriented perovskite crystals with its basal plane parallel to the substrate surface were formed.

3. Lasing characteristics of perovskite crystals

For lasing measurements, a Ti:sapphire femtosecond pulsed laser (λ = 397 nm, 1 kHz repetition rate, 200-300 fs duration, spot diameter: 83 μm) beam was focused with an incidence angle of 60° with respect to the perovskite crystals. The emission from the waveguide edges was detected by a charge-coupled device spectrometer (Roper Scientific ST-133 series).

Figure 2 shows the excitation density dependence of photoluminescence (PL) spectra of the $CH_3NH_3PbBr_3$ crystals formed inside the channel-type waveguide with DFB gratings.

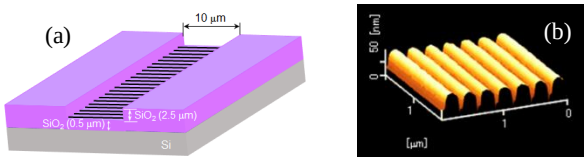

Fig. 1. (a) Schematic representation of a channel-type waveguide with DFB structure. (b) An AFM image of the DFB grating.

Above an excitation threshold of 47 μJ/cm^2, the PL spectra showed a clear linewidth narrowing of emissions at λ = 546 and 548 nm with a full width at half maximum of approximately 0.8 nm. This threshold value was one order of magnitude lower than that for amplified spontaneous emission (ASE) in the perovskite crystals formed inside channel-type waveguide without DFB gratings. With further increase in the excitation density, the emission linewidths were broadened, and then the lasing modes were obscured due to the high ASE intensities. These PL characteristics can be explained that as the excitation density is increased, the DFB lasing first arises in the vicinity of the interface between the DFB grating and perovskite crystal owing to the effective DFB gain. Above the DFB lasing threshold, the stimulated emission gain is predominant in the bulk region of the perovskite crystal apart from the interface.

Next, the DFB lasing modes were investigated using finite-differential time-domain (FDTD) calculations (two-dimensional simulations) for $CH_3NH_3PbBr_3$ on the DFB structure with a 253 nm period. In the calculations, the material dispersions are contained as a two-Lorentzian model according to the prior work by Sasaki *et al* [15]. The two peaks in the calculated spectrum in Fig. 3 at λ = 545.5 and 548 nm in the calculations showed a good agreement with the lasing mode peaks in the experimental results. The FDTD simulation results show two standing waves dephased by $\pi/2$, corresponding to the two DFB modes.

4. Conclusions

The DFB lasing was achieved for $CH_3NH_3PbBr_3$ crystals grown inside channel-type waveguides with DFB gratings fabricated by the FIB lithography. The highly oriented $CH_3NH_3PbBr_3$ crystals embedded in the waveguide with the DFB gratings demonstrated second-order DFB lasing modes above a threshold excitation density of 47 μJ/cm^2 which was one order of magnitude lower than that in the non-DFB laser. Moreover, the FDTD calculations well reproduced the two emission peaks at λ = 545.5 and 548 nm corresponding to the DFB modes dephased by $\pi/2$ for the second diffraction order. The splitting two DFB peaks changed to a power-broadened ASE as the excitation density was increased far above the threshold.

Acknowledgements

This work was supported by Japan Society for the Promotion of Science (JSPS) KAKENHI (Grant No. 19H02172). We thank Mr. S. Katao, Nara Institute of Science and Technology for his help with the XRD measurements.

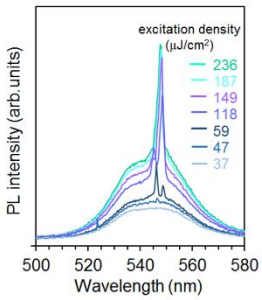

Fig. 2. Excitation density dependence of PL spectra of $CH_3NH_3PbBr_3$ crystals formed inside a waveguide with DFB gratings.

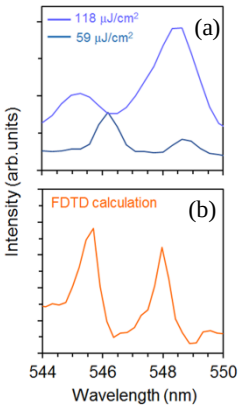

Fig. 3. Measured lasing spectra (a) and calculated spectrum (b).

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